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Long-range transport and airborne measurements of VOCs using proton-transfer-reaction mass spectrometry validated against GC-MS-canister data during the ASIA-AQ campaign

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Abstract

Volatile organic compounds (VOCs) play a critical role in atmospheric chemistry, contributing to the formation of ozone, secondary $PM_{2.5}$ production, and global warming. This study investigates the spatial distribution and long-range transport dynamics of VOCs over South Korea, using airborne observations conducted during the 2024 Asian Air Quality campaign. VOC concentrations were measured in urban, industrial, and long-range transport scenarios using proton transfer reaction-time-of-flight mass spectrometry (MS) and gas chromatography–MS with canister sampling. The results demonstrate that benzene shows significant vertical and spatial dispersion during long-range transport due to its intermediate atmospheric lifetime, which allows it to persist and impact downwind air quality. Additionally, Chlorinated VOCs, such as 1,2-dichloroethane and 1,2,4-trichlorobenzene, display transport behaviors. Their relatively consistent concentrations during long-range transport emphasize the influence of industrial activities, including coal combustion and petrochemical processes, as major sources. Elevated levels of chlorinated VOCs were primarily associated with emissions from industrial regions in Chungnam, while aromatic VOCs were predominantly linked to urban traffic emissions. These findings underscore the need for international cooperation to combat transboundary pollution and highlight the importance of comprehensive air quality management strategies that address both urban and industrial emission sources. This study provides essential insights into the atmospheric behavior of VOCs and emphasizes the need for targeted policies to effectively regulate air pollution.

1. Background

Volatile organic compounds (VOCs), identified as hazardous air pollutants, are released from a variety of sources including automobiles, gas stations, and industrial facilities (Lv *et al* 2021, Yuan *et al* 2022, Muda *et al* 2024) VOCs are pivotal in atmospheric chemistry, acting as key precursors to tropospheric ozone (O_3) and secondary $PM_{2.5}$ (Zheng *et al* 2021, Chen *et al* 2023). Moreover, VOCs contribute to global warming and odor pollution, thus impacting air quality across multiple dimensions (Kansal 2009, Murphy *et al* 2014, Unger 2014, Lee *et al* 2023). Some VOCs with high chemical stability persist in the ambient atmosphere, exacerbating global warming along with other greenhouse gases (Hodnebrog *et al* 2018, David and Niculescu 2021, Yu and Li 2021). These properties highlight the dual role of VOCs as both

local air pollutants and major contributors to global atmospheric changes.

Given the influence of atmospheric circulation and its geopolitical context, South Korea is particularly vulnerable to transboundary air pollution originating from neighboring Northeast Asian countries (Oh *et al* 2023, 2024). Long-range transport of pollutants, especially from China, often leads to episodes of high-concentration pollution within South Korea. This situation has prompted vigorous monitoring and modeling efforts to evaluate the impacts of these transboundary effects (Lee *et al* 2019, Kumar *et al* 2021). Significantly, VOCs transported over long distances can transform into secondary $PM_{2.5}$ within South Korea, further compounding fine particulate matter levels. Addressing such intricate pollution challenges requires a thorough understanding of domestic emission sources and their impacts, as well as precise identification of transboundary pollutants and their transport pathways—essential elements for both scientific research and effective policymaking (Oh *et al* 2020).

The 2016 Korea–United States Air Quality (KORUS-AQ) study has provided critical insights into VOC emissions in South Korea. Conducted jointly by South Korea's Ministry of Environment and the National Aeronautics and Space Administration (NASA), this airborne observation campaign revealed high concentrations of benzene and 1,3-butadiene over the Daesan petrochemical complex in Chungnam Province (Fried *et al* 2020, Crawford *et al* 2021). Low-altitude air sampling over Seoul, using NASA's Douglas (DC-8) research aircraft, identified isoprene, toluene, xylenes, and ethene as significant contributors to OH reactivity in the area. The study found that ozone formation was highly sensitive to aromatic VOCs and anthropogenic alkenes (Simpson *et al* 2020). Moreover, analyses of carbonyl sulfide (OCS) and carbon monoxide (CO) have pinpointed coal combustion as the primary source of OCS emissions, along with contributions from long-range transport of pollutants such as OCS, methyl chloride (CH_3Cl), 1,2-dichloroethane, ethyl chloride, and Halon-1211 from China (Barletta *et al* 2009). Recent research indicates that long-distance transport of VOCs, including 1,2-dichloroethane, 1,4-dichlorobenzene, and chlorobenzene, strongly correlates ($r^2 = 0.70$) with $PM_{2.5}$, highlighting their role in secondary organic aerosol formation (Oh *et al* 2024).

Despite the critical relevance of studying VOC precursors, especially those transported over long distances, challenges in conducting three-dimensional measurements remain significant. It is essential to supplement surface-level observations of domestic VOC emissions with three-dimensional atmospheric monitoring of upper air layers to better comprehend the role of long-range transport. In this study, a 1900D aircraft equipped with the proton transfer reaction-time-of-flight MS (PTR-ToFMS) system and the canister sampling system was employed during the Airborne and Satellite Investigation of Asian Air Quality (ASIA-AQ) campaign. The goal was to quantify VOC concentrations, analyze their spatial distributions, and trace transport pathways from (1) the Seoul metropolitan area, (2) major industrial emission sources, and (3) long-range transported VOCs.

2. Methods

2.1. Airborne observations during the ASIA-AQ campaign

Airborne observations were conducted using the Hanseo B-1900D aircraft during the 2024 ASIA-AQ campaign, an international collaborative field study that addressed regional air quality issues through the integration of satellite observations, ground-based monitoring, and modeling (www-air.larc.nasa.gov/missions/asia-aq/) (Park *et al* 2020). The Hanseo B-1900D aircraft, capable of a maximum cruising speed of 526 km h^{-1}, an operational ceiling of 7620 m, and a range of 2289 km, was equipped to accommodate up to 19 passengers (Wong *et al* 2024). It was outfitted with scientific instruments, including the PTR-ToF-MS system and the canister sampling system, both installed in the cabin behind the cockpit (figure S1). The PTR-ToF-MS system was centrally placed, followed directly by the canister system, with auxiliary equipment such as power supplies and pumps positioned towards the rear.

Throughout the campaign, the aircraft completed 16 research flights (RF16–RF31) from 7 February to 8 March 2024, predominantly investigating the Seoul metropolitan area, the West Sea, and industrial emission sources in Chungnam. The routes over Seoul metropolitan area and the West Sea were the most frequently monitored. During these flights, air samples were collected using four canisters per flight, resulting in a total of 56 collected canisters (table S1). Detailed specifications of the Hanseo B-1900D aircraft and its accompanying instrumentation have been documented in earlier studies (Wong *et al* 2024).

2.2. VOC measurement using PTR-ToF-MS

VOC measurements were conducted using the PTR-ToF-MS instrument (PTR-ToF-8000, IONICON Inc., Austria), which allows the analysis of a wide array of gaseous compounds with high temporal resolution. Samples were gathered via a gas inlet mounted on the aircraft's left side. This inlet incorporated two ports connected by 1/4-inch Teflon tubing, with the upper port used for chemical ionization MS and the lower port dedicated to PTR-ToF-MS sampling. A supplementary pump directed samples to the PTR front end at a flow rate of 5 l min^{-1}, and exhaust was channeled to a cabin exhaust line to uphold stable pressure. The

inlet was heated to 50 °C, with downstream components maintained at 80 °C to ensure their stability.

Pre-flight calibration was conducted using a standard gas mixture that included target compounds at specific concentrations of 0, 12, 23, 34, and 46 ppb, such as benzene, toluene, and styrene. Measurements were analyzed in real-time with a response time of around 1 s. Yuan *et al* (2017) noted potential interference between hydrated products of VOCs with the same nominal mass. However, studies (e.g. Permar *et al* 2021, Coggon *et al* 2024) found no significant interference from ethylbenzene or xylenes to toluene (m/z 93), though ethylbenzene fragmentation may affect benzene (m/z 79). Additionally, low protonation efficiency was noted for naphthalene, phenanthrene, as well as certain aldehydes and ketones. Compounds such as methanol and hydrogen sulfide posed quantification challenges due to uncertainties in calibration and injection. Semi-VOCs with low vapor pressure, including phthalates and organophosphates, were ineffectively detected (Yuan *et al* 2017, Majchrzak *et al* 2018, Zhang *et al* 2022). Consequently, PTR-ToF-MS measurements were validated through comparison with GC-MS analyses of canister samples to produce quantitative data.

2.3. VOC sample collection and analysis using GC-MS-canister

Air samples were collected using 6 l SilcoCan canisters (Restek, France) across 16 research flights. The canisters were evacuated to a vacuum state before being filled with air samples for 30 s during each flight, with subsequent analyses conducted within 48 h to minimize sample degradation. This sampling duration facilitated a direct comparison with PTR-ToF-MS measurements. The samples were then transferred to low-temperature adsorption tubes (TD100-xr, Markes International, UK) with an internal standard of toluene-D8. VOCs were quantified using GC (7890A, Agilent, USA) coupled with MS (5975C, Agilent, USA). A total of 30 VOCs, including benzene and toluene, were analyzed (table S2). The analytical protocols adhered to established methodologies previously described (Kim *et al* 2023, Choe *et al* 2024, 2025).

3. Results and discussion

3.1. Comparison of measurement techniques

Figure 1 displays scatter plots comparing measurements from PTR-ToF-MS and GC-MS-canister for toluene, benzene, and trimethylbenzene. The data points indicate a strong agreement between the two methods for these compounds. The slopes of the regression lines provide correction factors to calibrate PTR-ToF-MS data, helping to reduce systematic discrepancies and better align them with GC-MS-canister measurements. The focus on benzene, toluene, and trimethylbenzene from PTR-ToF-MS, as well as the eight chlorinated VOCs from GC-MS with canister sampling, was determined based on their validation as tracers for specific source identification. These compounds are widely recognized for their significance in urban and industrial emissions and their established associations with long-range transport and transboundary pollution, as highlighted in previous studies.

3.2. VOC concentrations across scenarios

Tables 1 and 2 outline VOC concentrations in five key scenarios: the Metropolitan area (Seoul and its surroundings), Chungnam industrial emissions (including South Korea's largest coal-fired power plant, identified by the Clean Air Policy Support System (CAPSS)) (Choi *et al* 2022), and three phases of atmospheric transport (Pre-, On-, and Post-Long-Range Transport) determined from flight tracks of benzene, toluene, and trimethylbenzene and their Impact on downwind air quality, supported by ERA5 and $PM_{2.5}$ Data (figure 4).

3.2.1. Aromatic VOCs

Table 1 highlights validated concentrations of three aromatic VOCs (toluene, benzene, and trimethylbenzene) based on calibrated PTR-ToF-MS data. In the Metropolitan area, toluene exhibits the highest concentration (3.71 ± 2.13 ppb), reflecting significant vehicular and anthropogenic emissions. Benzene levels (0.48 ± 0.17 ppb) and the elevated toluene-to-benzene ratio (7.73 ± 3.24) suggest a predominant influence from traffic-related emissions (Song *et al* 2021). While lower, trimethylbenzene levels (0.13 ± 0.04 ppb) are typical of urban environments. In the Chungnam emission area, the toluene-to-benzene ratio lowers to 2.07 ± 0.45, indicating reduced vehicular impact and increased industrial contributions. During atmospheric transport stages (Pre-, On-, and Post-Long-Range Transport), toluene and trimethylbenzene concentrations diminish, reflecting atmospheric dilution and chemical transformation, whereas benzene levels remain relatively stable owing to its moderate atmospheric longevity. The toluene-to-benzene ratio effectively differentiates between urban traffic and industrial emissions.

3.2.2. Chlorinated VOCs

Table 2 focuses on chlorinated VOCs (e.g. 1,2-dichloroethane and tetrachloroethylene) measured using GC-MS-canister analysis. These compounds, including 1,2,4-trichlorobenzene (1,2,4-TCB), 1,2-dichloropropane (PDC), and 1,2-dichloroethane, primarily originate from industrial processes such as chemical production and coal combustion (Barletta *et al* 2009). For instance, 1,2,4-TCB, a by-product of benzene chlorination, is used in dyes, pesticides, and solvents (van Wijk *et al* 2006, Shang

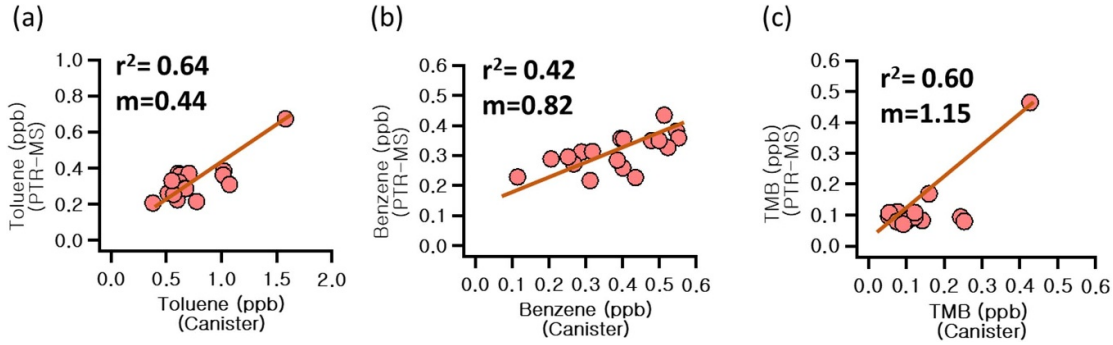

Figure 1. Scatter plots comparing PTR-ToF-MS and GC-MS-canister measurements for (a) toluene, (b) benzene, and (c) trimethylbenzene.

Table 1. Concentrations of toluene, benzene, trimethylbenzene, and toluene-to-benzene ratios during selected periods analyzed using PTR-ToF-MS calibrated with GC/MS-canister data.

VOCs (ppb) from PTR-ToF-MS	Metropolitan	Chungnam Emission	M.W.01[a]	M.W.02[b]	M.W.03[c]
Toluene	3.71 ± 2.13	0.56 ± 0.13	1.1 ± 1.18	0.7 ± 0.29	0.59 ± 0.14
Benzene	0.48 ± 0.17	0.27 ± 0.04	0.35 ± 0.1	0.38 ± 0.1	0.31 ± 0.07
Trimethylbenzene	0.13 ± 0.04	0.08 ± 0.02	0.09 ± 0.03	0.08 ± 0.01	0.07 ± 0.01
Toluene/benzene	7.73 ± 3.24	2.07 ± 0.45	3.14 ± 2.7	1.84 ± 0.7	1.9 ± 1.42

[a] Pre-Long-Range Transport Periods: Flights from Metropolitan to West Sea Areas.
[b] Periods of Long-Range Transport: Flights from Metropolitan to West Sea Areas.
[c] Post-Long-Range Transport Periods: Flights from Metropolitan to West Sea Areas.

Table 2. Concentrations of chlorinated VOCs during selected periods analyzed using GC-MS/canister.

VOCs (ppb) from GC-MS-Canister	Metropolitan	Chungnam emission	M.W.01[a]	M.W.02[b]	M.W.03[c]
Number of Canisters	8	4	12	20	8
1,2-dichloroethane	0.112 ± 0.037	0.122 ± 0.031	0.050 ± 0.024	0.125 ± 0.053	0.051 ± 0.020
1,2-dichloropropane	0.063 ± 0.021	0.078 ± 0.044	0.015 ± 0.011	0.043 ± 0.022	0.025 ± 0.020
1,1,2-trichloroethane	0.020 ± 0.021	0.019 ± 0.016	0.008 ± 0.005	0.010 ± 0.007	0.009 ± 0.012
Tetrachloroethylene	0.035 ± 0.019	0.079 ± 0.031	0.013 ± 0.008	0.021 ± 0.020	0.018 ± 0.012
1,2-dichlorobenzene	0.049 ± 0.061	0.069 ± 0.053	0.024 ± 0.020	0.039 ± 0.045	0.041 ± 0.042
1,3-dichlorobenzene	0.053 ± 0.068	0.104 ± 0.049	0.031 ± 0.025	0.053 ± 0.059	0.050 ± 0.045
1,4-dichlorobenzene	0.058 ± 0.066	0.079 ± 0.060	0.029 ± 0.024	0.038 ± 0.037	0.052 ± 0.052
1,2,4-trichlorobenzene	0.076 ± 0.091	0.109 ± 0.097	0.052 ± 0.045	0.071 ± 0.104	0.061 ± 0.066

[a] Pre-Long-Range Transport Periods: Flights from Metropolitan to West Sea Areas; RF20 (Feb.17), RF22 (Feb.23), RF23 (Feb.26).
[b] Periods of Long-Range Transport: Flights from Metropolitan to West Sea Areas; RF25 (Mar.02) to RF29 (Mar.04).
[c] Post-Long-Range Transport Periods: Flights from Metropolitan to West Sea Areas; RF30 (Mar.07), RF31 (Mar.08).

et al 2020). Oh *et al* (2024) studied a record-breaking air pollution episode in 2021, during which $PM_{2.5}$ exceeded 100 μg m^{-3}, and confirmed the inflow of pollutants from China to Korea using back-trajectory analysis and satellite observations. VOCs identified as transboundary pollutants from China included 1,2-dichloroethane (0.25 ppb), chlorobenzene (0.019 ppb), and 1,4-dichlorobenzene (0.006 ppb), with concentrations at least four times higher than those observed during non-event periods. Additionally, chlorofluorocarbons (CFC-113 and CFC-114), carbon tetrachloride, and 1,2-dichloroethane were identified as tracers of transboundary pollution (Simpson *et al* 2020). Its persistence in the environment is exacerbated by emissions from handling and combustion. The higher concentrations of tetrachloroethylene (0.079 ± 0.031 ppbv), 1,3-dichlorobenzene (0.104 ± 0.049 ppbv), and 1,2,4-trichlorobenzene (0.109 ± 0.097 ppbv) observed in the Chungnam region compared to the Metropolitan area suggest significant local emissions from industrial activities. These elevated concentrations indicate localized sources in Chungnam, such as industrial processes, contributing to regional air pollution and highlighting the need for targeted mitigation strategies. Notably, 1,2,4-TCB peaks during On-Long-Range Transport (0.071 ± 0.104 ppb),

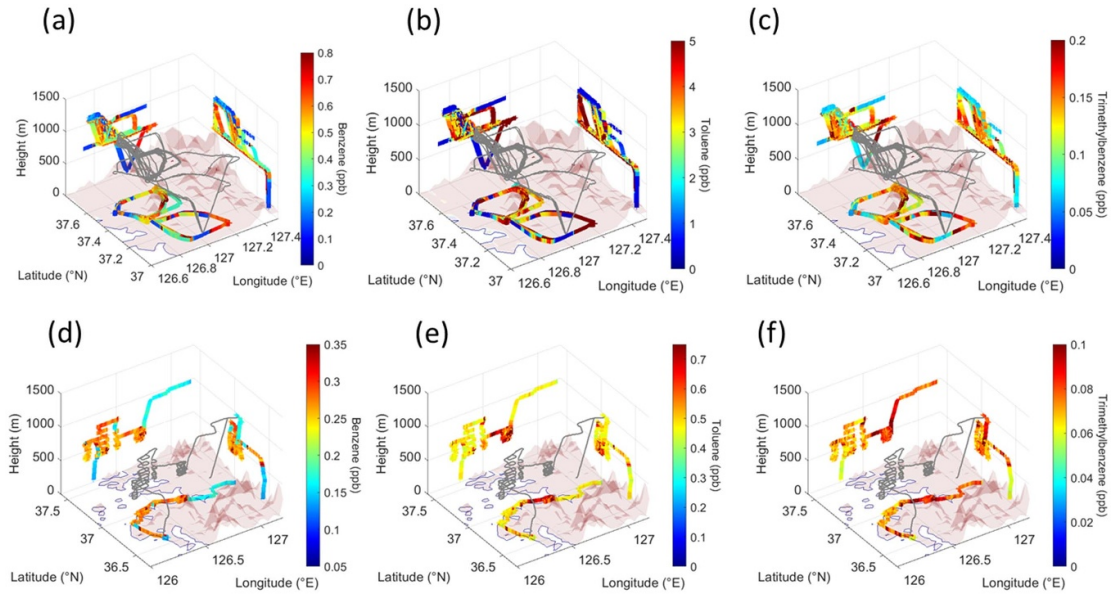

Figure 2. Flight tracks for (a), (d) benzene, (b), (e) toluene, and (c), (f) trimethylbenzene measured over the metropolitan emission areas (a)–(c) and the Chungnam emission area (d)–(f) in South Korea (gray color indicates flight tracks based on Hanseo B-1900D aircraft measurements).

highlighting the transport of industrial emissions across significant distances.

3.2.3. VOC dispersion and transport patterns

Figure 2 depicts flight tracks for benzene, toluene, and trimethylbenzene based on flight measurements over the Metropolitan area (figures 2(a)–(c)) and the Chungnam area (figures 2(d)–(f)). Solid circles indicate concentrations obtained from GC-MS-canister analysis. In the Metropolitan area, elevated benzene concentrations near ground level and within urban zones (latitude ∼37.0–37.8) suggest contributions from vehicular and local emissions. Toluene has a broader spatial distribution, extending across central urban zones and transportation routes, indicative of emissions from both vehicles and local industries. Meanwhile, trimethylbenzene concentrations are lower, concentrated near urban cores and along specific pathways.

In the Chungnam area, the higher benzene concentrations appear near industrial sites and coal-fired power plants (latitude ∼37.4, longitude ∼126.5), where vertical transport from smokestacks is observed. Toluene shows broader dispersion, while trimethylbenzene remains localized near emission sources.

3.3. Long-range transport of VOCs

Figures 3 and S3 illustrate flight tracks for benzene, toluene, and trimethylbenzene during Pre-, On-, and Post-Long-Range Transport phases. In the Pre-Long-Range Transport phase, elevated benzene levels (red regions) appear near emission sources at lower altitudes. Toluene is more widely dispersed at ground level, whereas trimethylbenzene remains localized. During the On-Long-Range Transport phase, benzene concentrations rise to higher altitudes (up to 2000 m), demonstrating vertical mixing and wider dispersion. Toluene maintains substantial concentrations both near the surface and at elevated altitudes, while trimethylbenzene shows limited transport. This is supported by ERA5 meteorological data and Air Korea $PM_{2.5}$ measurements, which confirm long-range transport events, with nationwide increases in $PM_{2.5}$ on March 3 (figure 4). In the Post-Long-Range Transport phase, there is a notable decrease in VOC concentrations due to dilution and chemical degradation. Benzene continues to be detected at mid-altitudes, whereas toluene and trimethylbenzene see substantial reductions, underscoring their shorter atmospheric lifespans. Figure 5 further emphasizes benzene's persistence and significant vertical dispersion during long-range transport, raising concerns about its impact on downwind air quality. Benzene's persistence and vertical dispersion during the On-Long-Range Transport period highlight its long-lived nature, with increased concentrations at 1000 m suggesting benzene-specific sources. The low toluene/benzene ratio (1.84) supports differing emission contributions compared to toluene, which reflects strong local sources.

These results highlight the distinct chemical signatures of urban and industrial emissions, the persistence of chlorinated VOCs, and the atmospheric behavior of benzene during long-range transport, underscoring the importance of understanding these dynamics for effective air quality management strategies.

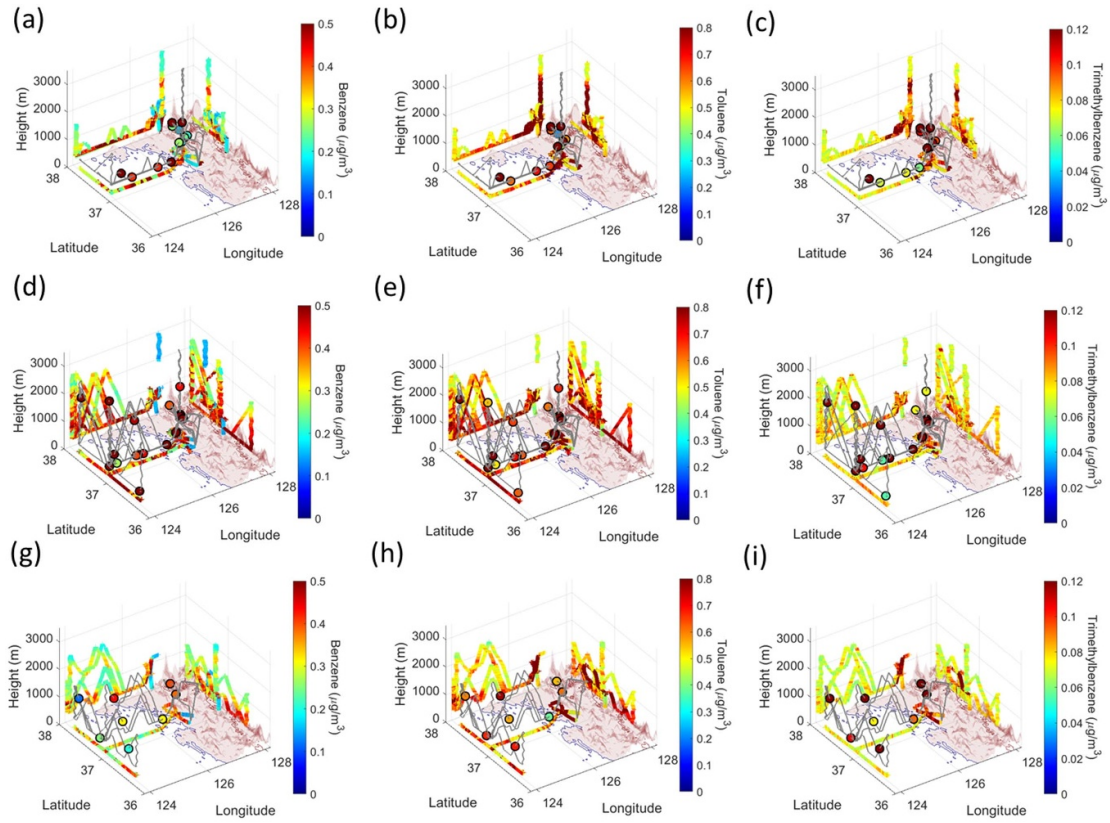

Figure 3. Flight tracks of benzene, toluene, and trimethylbenzene during different transport periods in South Korea. (a), (d), (g) benzene, (b), (e), (h) toluene, and (c), (f), (i) Trimethylbenzene for Pre-Long-Range Transport ((a)–(c): RF20, RF22, RF23), Long-range transport ((d)–(f): RF25–RF29), and post-long-range transport ((g)–(i): RF30, RF31) from metropolitan to West Sea Areas. Solid circles represent concentrations analyzed using GC-MS-canister.

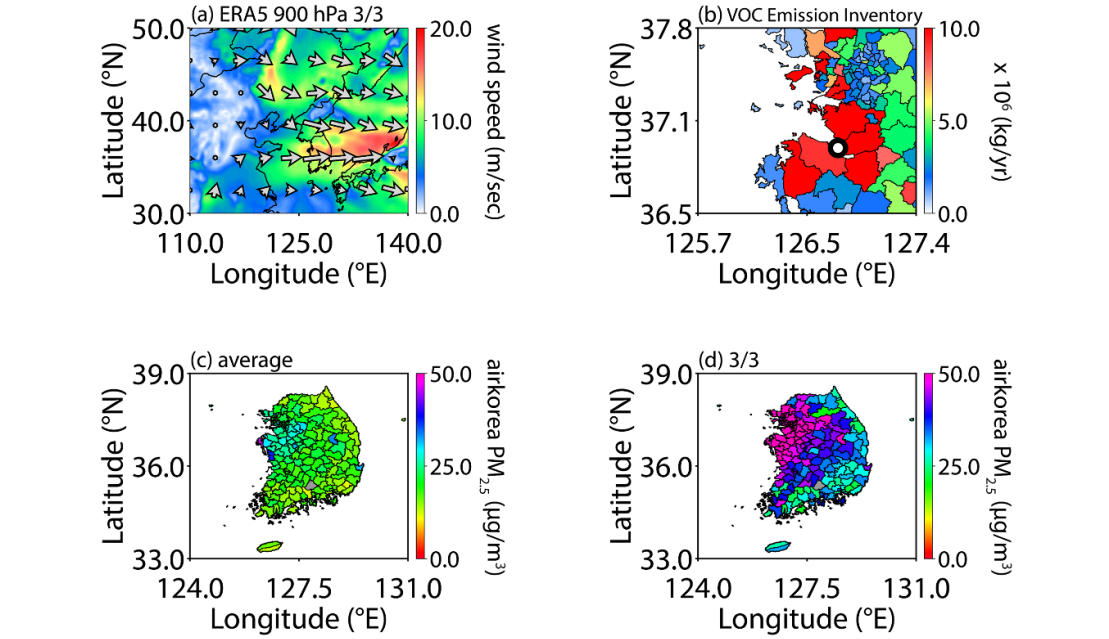

Figure 4. (a) Wind field data (ECMWF's ERA5, 900 hPa) on March 3, (b) PM emissions in the Chungnam area from the clean air policy support system (CAPSS), and (c) National $PM_{2.5}$ concentrations from AirKorea (www.airkorea.or.kr/web/) during the entire period and long-range transport period on March 3 in South Korea.

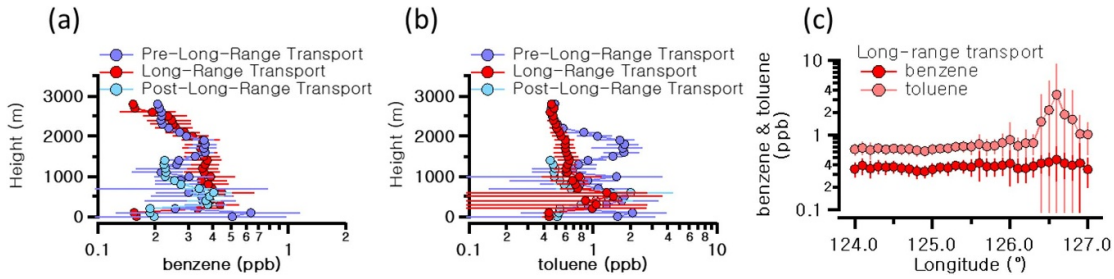

Figure 5. Vertical profiles of (a) benzene and (b) toluene concentrations during different phases of long-range transport and (c) spatial variation of benzene and toluene concentrations along a longitudinal gradient (124°E–127°E).

4. Conclusion

This study highlights the critical role of concentrations in the Metropolitan and Chungnam emission areas, along with long-range transport, in shaping the atmospheric distribution and persistence of benzene and chlorinated VOCs, with significant implications for air quality and public health. The elevated concentrations of benzene at higher altitudes during these periods underscore its persistence and vertical dispersion, particularly during the On-Long-Range Transport period, highlighting its long-lived nature. The increased concentrations at 1000 m suggest benzene-specific sources, with the low toluene/benzene ratio indicating differing emission contributions compared to toluene. This persistence allows benzene to adversely influence air quality in both source regions and distant downwind areas, raising concerns about its extensive role in regional and global air pollution. Chlorinated VOCs, such as 1,2-dichloroethane and 1,2,4-trichlorobenzene, display transport behaviors. Their relatively consistent concentrations during long-range transport emphasize the influence of industrial activities, including coal combustion and petrochemical processes, as major sources. The prevalence of chlorinated VOCs in the Chungnam industrial area and during transportation stages further highlights their strong link with industrial emissions rather than urban traffic sources. These observations necessitate the development of integrated air quality management strategies that accommodate both local and transboundary sources of VOCs. The enduring presence of chlorinated VOCs during transport underscores the importance of continuous monitoring of industrial activities and the enforcement of stricter emissions controls in source regions to minimize their long-range atmospheric impacts. Overall, this study offers essential insights into the interaction between local emissions, atmospheric processes, and transboundary transport, providing valuable guidance for crafting air quality policies and conducting environmental research aimed at reducing VOC-related pollution.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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